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INTEROFFICE CORRESPONDENCE

Date:

January 8, 1993

To:

R. L. Nitschke, MS 3960

From:

D. W. Rhodes, MS 3908 Devichele

Subject: EVALUATION OF RECORDS FOR WASTE GENERATED AT THE IDAHO CHEMICAL PROCESSING PLANT (ICPP) AND DISPOSED AT THE RADIOACTIVE WASTE

MANAGEMENT COMPLEX (RWMC) DURING THE PERIOD OF 1960-1983 - DWR-01-93

Attached is an evaluation of waste generated at the Idaho Chemical Processing Plant, and buried at the Subsurface Disposal Area during the period 1960-1983, as per your request. Emphasis was on waste that may be considered non-mobile, either due to the nature of the waste itself, or because of the integrity of the container.

Only radionuclides with a half-life greater than five years were considered.

If you have any questions or need additional information, please contact me at 526-6998.

knt

Attachment: As Stated

cc: C. J. Barnard, MS 3514 J. L. Butler, MS 3908 ARDC File, MS 3904 Central Files, MS 1651 D. W. Rhodes File

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WASTE DISPOSED TO THE RADIOACTIVE WASTE MANAGEMENT COMPLEX (RWMC) FROM THE IDAHO CHEMICAL PROCESSING PLANT (ICPP) (1960-1983)

Generation of Waste

The Idaho Chemical Processing Plant (ICPP) is an operating facility at the Idaho National Engineering Laboratory (INEL) that processed military and test reactor fuels, as well as fuel materials from small-scale special experimental projects. In 1992, the processing of fuel was discontinued permanently. In addition to processing fuel, high-level radioactive waste, that is produced by processing fuel, is converted to a granular solid in a fluidized bed calciner for long-term storage in underground stainless steel bins.

Radionuclides resulting from the chemical processing of expended nuclear fuels, decontamination of process cells, and concentration of solutions in the Process Equipment Waste (PEW) evaporator are routed to 300,000 gallon underground waste storage tanks for storage and eventual processing in the fluidized bed calciner. Although the ICPP processed fuels that produced hundreds of thousands of curies of radionuclides, nearly all of these radionuclides remained at the ICPP, contained in the waste storage tanks or the calcine storage bins.

Operations that produce solid waste that is disposed to the Radioactive Waste Management Complex (RWMC) include, replacement of obsolete equipment, wipe down of fuel shipping casks, disposal of fuel end boxes, removal of sludge from the fuel storage basin, replacement of dry air filters, discard of anti-C clothing, and custom processing of special materials in the multicurie cell.

Characteristics of Waste

Based on information obtained from the Radioactive Waste Management Information System (RWMIS) database, approximately 2.342E+04 cubic meters of solid waste containing 4.724E+05 curies of radionuclides were transported from the ICPP to the RWMC in the period 1960-83. The radionuclides with half-lifes greater than five years that were contained in this waste are shown in the following table along with the number of curies, and the number of shipping records that listed the radionuclide.

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<u>Radionuclide</u>	<u> Half-life (yrs)</u>	No. of Records	No. Curies
Cs-137	30.17	650	1.493E+04
Sr-90	29.1	603	3.877E+03
Sr-Y-90	29.1	5	1.549E+03
Co-60	5.27	152	2.006E+05
MFP	-	3542	9.981E+04
MAP	-	43	2.308E+04
UN-ID-BG	-	25	1.150E+03
Eu-152	13.48	60	3.955E+02
Eu-154	8.59	262	3.674E+02
Ra-226	1.60E+03	2 1	1.002E-01
Np-237	2.14E+06	1	4.200E-06
Th-232	1.4E+10	11	1.137E-07
U-233	1.59E+05	1	5.740E-05
U-234	2.46E+05	40	1.635E-01
U-235	7.04E+08	104	1.200E-02
U-236	2.342E+07	39	4.019E-03
U-238	4.47E+09	90	2.009E-01
Pu-238	87.7	51	1.068E+00
Pu-239	2.42E+04	76	9.027E-01
Pu-240	6.56E+03	67	2.354E-02
Pu-241	14.4	19	1.503E+00
Pu-242	3.75E+05	19	8.892E-05
Am-241	432.7	1	2.100E-04

For the purposes of this evaluation, the term MFP (mixed fission products), which was listed on 3,542 shipment records, was considered to include 10% Cs-137 and 10% Sr-90. The basis for this is the document, EGG-WM-9857 (2). In this document, CPP personnel responded to a questionnaire, which asked them to provide their best estimate of the radionuclide breakdown for the MFP in CPP waste. These are probably reasonable values (probably at least within a factor of 2), because a computer program to calculate the fission product activity in "typical" depleted fuel after 1 and 30 years cooling, Letter from R. L. Tromp to D. W. Rhodes (Tro-07-90), produced values of 6.6% and 23.68% for Sr-90 for 1 year and 30 years cooling periods respectively, and 7% and 25.5% for Cs-137 for 1 and 30 years cooling periods respectively.

For the entries listed as Sr-Y-90, the value was halved, because the Y-90 was considered to be in secular equilibrium with Sr-90.

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FUEL END BOXES

The RWMIS printouts show that the predominant radionuclide (42% of the total curies from CPP) is the activation product, Co-60. This radionuclide was listed on 152 shipment records, but 99% of the total Co-60 was contained in 13 shipments of EBR-II fuel end boxes from the CPP-603 fuel storage facility. The other radionuclides listed as being in the fuel end boxes are, CO-58, Cr-51, Mn-54, and MAP. These are shown in Table 2. All totalled, they account for approximately 69% of the total curies disposed to the RWMC from the ICPP in the time period 1960-1983.

The EBR-II fuel rods are clad in stainless steel, and the fuel assembly is also clad in stainless steel. The Co-60 and other activation products are formed by neutron activation of the metallic elements that make up the stainless steel. Prior to processing the fuel, the end boxes, which do not contain any uranium, are sawed off and disposed as waste. The Co-60 is an integral part of the stainless steel fuel end boxes, and as such would be essentially non-mobile during storage.

The individual shipments of fuel end boxes from the ICPP averaged about 15,000 curies per shipment. This is probably a reasonable value, because shipments of fuel end boxes from the Naval Reactor Facility (NRF) were sometimes higher than this by more than a factor of 10.

Table 2. Total Amount of Radionuclides in Fuel End Boxes from the ICPP (from the RWMIS Database).

<u>Radionuclide</u>	Total Amount (Curies)
Co-58	3900
Co-60	198250
Cr-51	78975
MAP	23075
Mn - 54	20800
Total	325000

SLUDGE CASKS

In the mid-1970's, sludge that had accumulated on the floor of the CPP-603 fuel storage basin facility for 20+ years was removed, and disposed to the RWMC. This sludge consisted of wind blown silt from the outside, and corrosion products from fuel cladding and underwater fuel storage and handling equipment in the basin. The chemical composition of the sludge is shown in "Table 3." The sludge contained significant quantities of radionuclides that had leaked into the water from stored fuel elements and fuel scrap cans since startup of the facility in 1952.

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The sludge was vacuumed into a large underground, stainless steel collection tank, then transferred into large concrete casks (see attached sketch "Figure 5"), where it was allowed to settle, the supernatant liquid decanted, and urea-formaldehyde added to solidify the sludge. Finally, concrete was poured into the opening to form a permanent plug.

Some 42 sludge casks were transported to the RWMC and buried in Pit 16. Minor cracks were observed on a few of the casks before they were transported to the RWMC. This was believed to be caused by the freezing of liquid that had entered the space between the steel liner and the concrete wall of the cask during the filling operation. These cracks were patched with Alemite Rubber Sealing Compound, and the top of each cask was cleaned and coated with Alemite Plastic Armor epoxy. This minor cracking was believed not to affect the physical integrity of the casks. Apparently, this was true, because they were loaded on a low-boy trailer and transported to the RWMC without incident.

Sampling and Analysis

Since the sludge contained a small amount of U-235, there was some concern about the possibility of a criticality when the sludge was concentrated by transferring it to the collection tank.(1) A criticality limit of 0.145 wt % on a dry weight basis was used as the limit. Extensive sampling and analysis was done before the sludge was vacuumed from the basin floor. If the standard deviation for the U-235 content of all samples in a given area was > 0.04 wt%, three additional samples were taken and analyzed for uranium. The results of this second sampling were used to determine if a special procedure was needed.

The results of a radiochemical analysis of the sludge from each of the three fuel storage basins are shown in the attached Table 5; column A is the analysis that applies to the contents of the sludge casks. As seen in Table 5, the concentrations of the radionuclides from the three different basins varies by a factor of 4 for Ce-144, by a factor of 2 for Sr-90, and is essentially the same for Cs-137. Since the sludge was vacuumed from only one basin at a time, and was mixed thoroughly in the collection tank before sampling and subsequent transfer to the sludge casks, the concentration values for the individual radionuclides very likely varies by considerably less than a factor of 2.

TABLE 3

CHEMICAL MAKEUP OF SLUDGE (SOLIDS ONLY)*

(Wt%)

	DATE				
Cation '	May .70	Aug. 73	Aug. 76		
Al	10.0	10.0	1.5		
Ca	.1	.1	2.3		
Cr	. 2	.02	. 2		
Cu	.2	.02	.02		
Fe	5.0	6.0	35.0		
Mg	3.0	3.0	.1		
Mn	.1	.1	.3		
РЪ	. 2	2	.6		
Si	20.0	20.0	17.8		
Ti .	.1	.1	•3		
Zn	3.0	3.0	1.2		
Zr	.001	.001	.02		
Na			1.3		

^{*} See Table 5 for radiochemical composition Anion content is a mixture of oxides, carbonates, sulphates, oxalates, silicates, hydroxides, and bicarbonates.

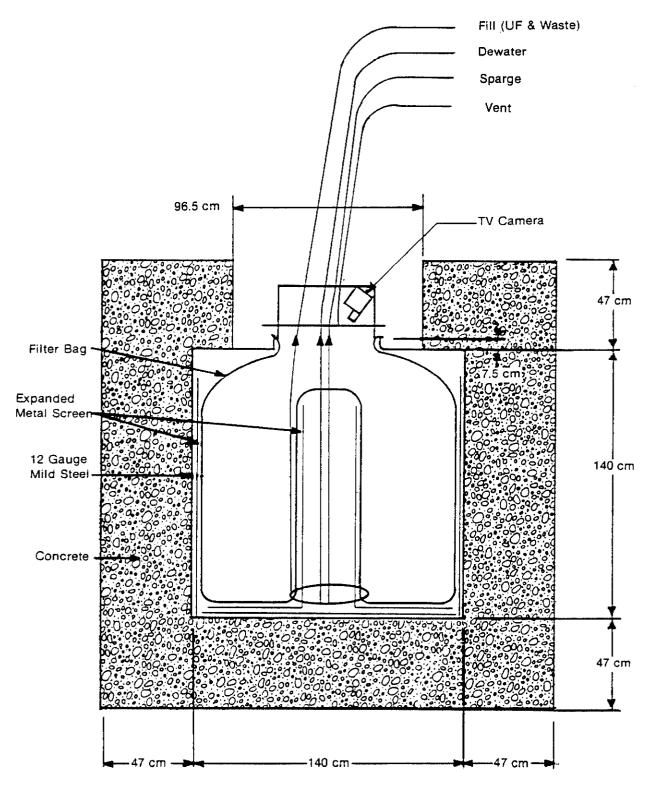


FIGURE 5. CONCRETE VAULT WITH STEEL LINER FOR SOLIDIFICATION AND STORAGE OF SLUDGE

TABLE 5 BASIN SLUDGE RADIOCHEMICAL ANALYSIS — μ Ci/gm Solids

	NORT	H BASI	N	мі	DDLE BAS	IN	S	OUTH BASI	
Isotope	A	В	C	A	В	C	A	В	C
Sr-89	4,81			2.84			7.03	 .	
Sr-90	6.10			. 6.77			12.3		
Ce-144	561 . 5	52.2	6.70	226	343	4.81	937	522	130
Eu-152	54.5	*** •••	-	20.0	ين وو ت	.———	9.57		
Eu-154	58.2			25.7			6.70		
Co-60	33.4 5	51.1	19.9	9.47	16.3	4.38	5.38	15.5	4.11
Cs-137	25.9 1	9.3	5.35	25.4	19.5	5.57	27.2	39.5	i5.9
Eu-155	16.1			6.15			55.5	======	
Nb ·95	17.7	6.81	•562	6.76	50.8	.098	4.50	13.7	6.57
Zr-95	12.9	5.00		4.93	35.9	.303	3.06	7.73	.67
Ru-106	7,00	3.78	1.31	3.61	8.62	1.00	2.14	58.4	25.7
Ce-141	6.34	7.03		2.43	36.5	.045	18.1	6.22	
Sb-125	2.39	.171		•723	Alate and a second		•334	1.08	•93
Cs-134	1.74	.384	.210	1.11	•557	.087	.933	.608	.26
La-140	•962			•428			.361		
Mn-54	.743	.565		.335	1.10		.356	38.9	
Co-58		.403	Noncomp event	*****	.695			28.4	
Ru-103	Companies of the	1.40		March relief WHITE	10.4			.689	

A = Aug. 76 samples, 1 per basin

⁼ Aug. 73 samples, 5 per North and Middle Basins, 9 per South Basin

C = Taken prior to mid 1971 - as reported in ACI-105

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Discussion

The total curies of each radionuclide summed for all of the sludge casks is shown in "Table 1". These values were obtained from the RWMIS Database. Overall, the predominant radionuclide in the sludge casks was Cs-137, even though the predominant radionuclide in the sludge on the basin floor was Ce-144 (Table 5). The Ce-144 sorbed readily on the sludge particles and thus became concentrated. The highly soluble Cs-137, on the other hand, remained in the basin water. However, the water in the storage basin is circulated continuously through an inorganic ion exchanger, which when exhausted, is discharged to the sludge collection tank. This ion exchanger material then becomes a part of the sludge. This accounts for the fact that Cs-137 is the predominant radionuclide in the sludge that was transferred to the sludge The curies of Sr-90 are low relative to the Cs-137 and Ce-144, because it is removed from the basin water by an organic ion exchanger. This ion exchanger is regenerated, and the spent regenerant is routed to the plant evaporator, where it is concentrated and stored in the 300,000 gallon highlevel radioactive waste storage tanks for eventual calcination.

This project produced approximately 48,150 liters of sludge weighing approximately 195,000 kilograms(1). Based on values obtained from the RWMIS Database, this sludge contains approximately 5% of the total curies disposed from the ICPP to the RWMC during the period 1960-83. This amounts to approximately 16% of the "non-mixed activation product curies", as well as about 77% of the total plutonium. The waste in these casks is important, because the radionuclides contained in the sludge can be considered non-mobile for the near term because of the container, and will not be available for leaching until the 18-inch thick walls of the concrete casks disintegrate.

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Table 1. Total Amount of Radionuclides Contained in Sludge Casks from the ICPP Fuel Storage Basin (from the RWMIS Database)

<u>Radionuclide</u>	Total Amount (Curies)
Ce-141	31.2
Ce-144	4314.0
Co-60	74.0
Cs-134	429.4
Cs-137	11491.7
Eu-152	236.4
Eu-154	231.5
Eu-155	78.5
MFP	105.6
Nb-95	68.5
Pr-144	4314.0
Pu-238	0.8087
Pu-239	0.3768
Pu-240	0.0098
Pu-241 Pu-242	1.5030
Rh-106	0.0001
Ru-106	75.4 75.4
Sb-125	73.4 6.2
Sr-90	1478.7
U-234	0.2327
U-235	0.0056
U-236	0.0037
U-238	0.0037
Y-90	1478.7
Zr-95	46.7
Total	24539.2
IVVI	LTUUJ.L

Summary

When the radionuclides contained in the fuel end boxes and the sludge casks are totalled, they account for about 74% of the total radionuclides disposed to the RWMC from the ICPP in the period 1960-83. This information is summarized in the following tables.

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Table 4. Quantity of Radionuclides in Fuel End Boxes and Sludge Casks

<u>Waste</u>	<u>Curies</u>	% of Total	Burial Location
Total Waste	4.724E+05	100	RWMC
Fuel End Boxes Sludge Casks Total	3.250E+05 2.360E+04 3.486E+05	69 <u>5</u> 74	Trench-57 Pit 16

Table 6. Distribution of Long-lived (greater than five years) Radionuclides in Fuel End Boxes and Sludge Casks from the ICPP.

<u>Waste Type</u>	<u>Radionuclide</u>	<u>Curies</u>	<pre>% of Total from CPP for Listed Radionuclide</pre>
Fuel end boxes	Co-60 MAP	1.9825E+05 2.3075E+04	99 100
Sludge Casks	Cs-137 Sr-90	1.149E+04 1.479E+03	67 32
	U-234 U-235 U-236 U-238 Total	1.327E-01 5.600E-03 3.700E-03 9.000E-04 1.429E-01	38
	Pu-238 Pu-239 Pu-240 Pu-241 Pu-242 Total	8.807E-01 3.768E-01 9.700E-03 1.503E+00 1.000E-04 3.4973E=00	77
	Eu-152 Eu-154 Total	2.364E+02 2.315E+02 4.679E+02	61

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OTHER WASTE

General

Wastes containing the other 26% of the radionuclides consist of a very large variety of waste materials. These include wood, metal, paper, anti-C clothing, glass, plastic, dirt, rags, concrete, rubber, insulation, filter aid, lead, and cardboard. These materials are present in various forms such as boxes, pipe, bottles, broken glass, gloves, some small amounts of liquid absorbed in gypsum or vermiculite, cans, tanks, wire, barrels, valves, and numerous other discarded items. In general, the radionuclides (1.23E+05 curies) are considered to be potentially mobile, i.e., subject to leaching if contacted by large amounts of water.

Lead

Approximately 45,000 pounds of lead from the ICPP were disposed to the RWMC. This lead was essentially all in the form of lead bricks and lead sheets, and as such is not considered to be mobile. A little more than half of this (24,000 pounds) was disposed to Pit 15 on the same date (May 30, 1978).

Decontamination Chemicals

Chlorinated hydrocarbons such as Methachlor and Oakite Swift were used in the decontamination of fuel shipping casks at the CPP-603 fuel storage facility. The procedure consisted of wiping the empty casks with a rag containing the hydrocarbon. The rag was then discarded to waste and eventually was disposed to the RWMC.

There are no records showing the quantity of these materials that was used. However, one of the operators (L. W. Madsen), who was involved in this operation for many years, estimated that they used approximately 1.5 gallons per month up through 1978. Use of these materials was discontinued after that time. Thus the total amount used for the period 1960-1978 would be:

The reliability of this estimate is not known. The amount disposed to the RWMC would be affected also by the fact that some of this solvent would evaporate from the rags while stored in open cardboard boxes prior to disposal.

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References

- 1. ICP-1195- Removal and Disposal of Radioactive Sludge from the Fuel Storage Basin at the Idaho Chemical Processing Plant, G. W. Hoech. and D. W. Rhodes, June 1979.
- 2. EGG-WM-9857- Analysis of the Low-Level Waste Radionuclide Inventory for the Radioactive Waste Management Complex Performance Assessment, L. E. Plansky and S. A. Holland, February, 1992.